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Exposure Analysis of Bisphenol A in Surface Water Systems in North America and Europe

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This study was conducted to develop a statistical understanding of exposures to bisphenol A (BPA) in aquatic environments in North America and Europe. Concentrations of BPA have been reported by 89 investigations published between 1997 and 2007. On the basis of an analysis of weighted observations (n = 1068 and 848 for North America and Europe, respectively), BPA was reported at concentrations above the detection limit in 20–51% of freshwater samples. Median BPA concentrations for fresh surface waters for North America and Europe were 0.081 and 0.01 μg/L, respectively, while 95th percentiles were 0.47 and 0.35 μg/L, respectively. In contrast to fresh surface waters, only limited data are available for sediments and less for marine ecosystems. For freshwater sediments in North America (n = 71), the median and 90th percentile concentration (the 95th percentile was not calculable) were 0.6 and 3.4 ng/g-dw, respectively, while the median and 95th percentile concentration in Europe (n = 249) were 16 and 256 ng/g-dw, respectively. To assess the potential ecological significance, we compared exposure concentrations with available regulatory criteria. The results suggest the frequency of locations in which concentrations are likely to cause adverse effects on aquatic ecosystems is low, with the exception of sediments collected from some highly urbanized and industrial locations.

Introduction

Bisphenol A (BPA, CAS registry number 80-05-7) is primarily used as an intermediate in the production of polycarbonate plastic and epoxy resins (*1*). BPA has also been reported to be used as a component of synthetic plastic materials such as polyvinyl chloride (PVC). The specific properties of polycarbonate plastic (e.g., lightweight, clear, and highly shatter-resistant) and epoxy resins (e.g., durable, chemically resistant, and good adhesion) have led to their use in a variety of consumer and industrial products.

Small amounts of BPA enter the environment from wastewater treatment plants, and the aquatic compartment has been identified as the main compartment in which BPA may be found (*2*). BPA will partition to organic phases such as soils and sediments (log octanol–water partition coefficient of 3.4); however, an appreciable fraction of BPA will remain in the dissolved phase (*2*). BPA is considered readily biodegradable (*3*), and aerobic biodegradation is the dominant loss process for BPA in all media (*4*) except the atmosphere, where it is susceptible to rapid reaction with hydroxyl radicals (*1*). BPA may be removed to the extent of 91–98% in municipal wastewater treatment plants (*5*).

Measurements of BPA in various environmental media have been reported in numerous studies by government agencies and other researchers (Supporting Information). For many of these studies, BPA is one of a long list of analytes, while other studies have focused on measurements of BPA only. Many of the studies are described as national monitoring programs, while others have the objective of characterizing concentrations in localized areas of suspected contamination. Several recent papers have reviewed concentrations of BPA in the environment (*6*, *7*), but none have critically reviewed the available data nor statistically characterized the concentrations of BPA in the environment.

The objective of this study was to statistically characterize the concentrations of BPA in North American and European aquatic environments. To develop an understanding of their potential ecological significance, we compared concentrations of BPA with available regulatory criteria.

Experimental Methods

Identification and Critical Evaluation of Studies. A literature search was conducted to identify environmental monitoring studies published between the early 1990s and 2007 that reported measurements of BPA in surface water and sediment in North America or Europe. Each study was critically evaluated for reliability using the review process described by Klečka et al. (*8*) that is generally based on internationally recognized guidance from Environment Canada (*9*) and the European Union (*10*). Studies categorized as "reliable" or "very reliable" were used in this investigation. An additional 11 studies considered "unassignable" but of sufficient quality to be retained because in all cases they were national studies conducted by European governments. Additional details for all studies and the criterion used in the evaluation are provided in the Supporting Information.

Statistical Treatment of Data. Data from the studies that passed the critical review were then subjected to statistical analysis. There were three issues that needed to be resolved: (1)] The concentration is reported as less than the detection limit for a large portion of the data set. For example, 80% of the measured concentrations of BPA in fresh surface water in North America are reported as less than the detection limit and 49% are less than the detection limit in Europe. (2) The detection limits are different for different studies. In North America, the most common detection limit is 1.0 μg/L; however, very few (0.6%) of the measured values exceed the highest detection limit (1.0 μg/L). (3) The data from the different studies must be combined to perform the statistical analysis. However, for some studies, only summary statistics are available (e.g., number of samples, frequency of detection, minimum, and maximum).

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To characterize nondetected concentrations, we considered several different approaches. Although substitution methods are commonly accepted by the regulatory community, the approach may obscure trends in a data set (*11*). The selected approach for the analysis of the BPA data set was the Kaplan–Meier (KM) method, which is recommended for data sets with the issues identified above (*11, 12*). The KM method is a nonparametric method, which assumes that all observations come from the same probability distribution. However, it does not require an assumption a priori of a specific distribution for the data. The KM method is especially suited to data sets with differing detection limits. Observations below the lowest detection limit are replaced by that detection limit if no other observations below that limit are available. Observations below higher detection limits are estimated from the distribution of measured values obtained from the studies with lower detection limits.

Some studies reported multiple measurements of samples collected from the same location. For the present analysis, each location was given the same weight in the calculation of the summary statistics, regardless of the number of times that location was sampled. Single unique observations were given a weight of one; multiple observations from the same location were given a combined weight of one. This weighting has the effect of reducing the total number of samples analyzed.

A number of studies reported only summary statistics. Corresponding authors were contacted and in most cases, the raw data were obtained. However, despite considerable effort, individual data points were not available for all, and these studies required special treatment. The method employed depended upon the summary statistics available for that study. For example, if the minimum, maximum, and median were available and all samples were above the detection limit, individual samples were imputed by equally spaced values in the cumulative of the log-normal distribution with the same minimum, maximum, and median values. These approaches allowed for generation of a data set, which could be collectively analyzed using the KM method.

The data were transferred to the software program S-plus 2000 (*13*) and the S-plus function "kaplanMeier" was used to calculate the "survival" probability for each concentration (i.e., the probability that the concentration in an arbitrary sample is equal to or higher than that concentration). If the observation was below the detection limit, a code indicating the data were censored was input. The S-plus function "MeanKM" was used to calculate median and 95th percentiles of the observed distribution. The percentiles were calculated by interpolation between the observed probability just under and just above these percentiles.

Results

Summary of Environmental Monitoring Studies. A total of 100 papers or reports, published between 1991 and 2007, were initially identified that contained environmental monitoring data for BPA in North American or European surface water and sediment. Following the data quality and analytical reviews, 11 papers were excluded (Supporting Information). The remaining 89 papers retained for analysis included 31 papers for North America and 58 papers for Europe (Supporting Information). Table 1 summarizes the geographic distribution of the available monitoring data, listing the number of studies and the number of samples available for each country. Many studies from North America and Europe characterized the sample locations as being downstream of wastewater discharges, receiving waters for industrial facilities, areas susceptible to contamination, urban waterways, or industrial ports.

Analytical detection limits varied from one study to another (Supporting Information). In general, the detection limits for water for the European studies were much lower (33 of 36 studies range from <0.0009 to 0.089 μg/L) than those for North America (15 of 24 studies have detection limits of 1.0 μg/L). The methods used in most of these studies were developed by the U.S. Geological Survey (*14, 15*) and were designed to detect multiple related analytes in a sample, likely sacrificing some of the analytical sensitivity.

TABLE 1. Geographic Distribution of Monitoring Data

	number of studies [number of samples][a]			
	surface water		sediment	
country	fresh	marine	fresh	marine
North America				
Canada	2 [5]	0	1 [55]	0
U.S.A.	24 [1548]	0	2 [25]	1 [14]
total	26 [1553]	0	3 [80]	1 [14]
Europe				
Austria	2 [306]	0	0	0
Belgium	1 [18]	0	0	0
Czech Republic	2 [21]	0	1 [37]	0
Denmark	2 [2]	0	1 [36]	1 [10]
France	1 [1]	0	0	0
Germany	8 [234]	3 [50]	5 [98]	0
Italy	5 [65]	3 [35]	2 [11]	0
Netherlands	3 [116]	2 [15]	1 [18]	0
Norway	1 [2]	0	3 [41]	2 [13]
Portugal	2 [207]	0	0	0
Spain	10 [72]	5 [26]	1 [6]	2 [15]
Sweden	0	0	1 [35]	1 [14]
Switzerland	2 [30]	0	0	0
U.K.	4 [55]	0	2 [65]	1 [22]
total	43 [1129]	13 [126]	17 [347]	7 [74]
overall total	69 [2682]	13 [126]	20 [427]	8 [88]

[a] Actual number of samples is larger than reported here as some studies do not report the number of samples; samples are not weighted. A paper may contain studies for multiple countries and/or media.

The proportion of samples where the concentration was reported as below the detection limit is large as shown in Table 2. Note that Table 2 presents the total number of weighted observations. Surface water in North America has the largest proportion of samples reported as less than the detection limit (80%, 852 of 1068 weighted observations) as compared to 49% for Europe (415 of 848 weighted observations). In general, between 30% and 79% of the sediment concentrations were reported as below the detection limit.

Surface Waters. Table 2 summarizes the results of the statistical analysis of BPA in surface water for each region, combining the data from all of the studies. Only a small number of samples of freshwater were imputed for the calculations (1 and 10 for North America and Europe, respectively). The estimated observations fit a log-normal distribution and match the summary statistics available for that study. It was not necessary to estimate observations for the marine water calculations. The median concentration in freshwater in North America is higher than that in Europe (0.081 and 0.01 μg/L, respectively); however, the 95th percentile concentrations are similar (0.47 and 0.35 μg/L for North America and Europe, respectively). Although the data are more limited, the median concentration of BPA in marine waters (available only for Europe) is about six times lower than that observed in fresh surface water (0.0016 versus 0.01 μg/L).

The distribution of BPA concentrations in North American and European fresh surface waters are illustrated in Figures 1 and 2 (additional distributions are presented in the Supporting Information). The left and right panels of Figures 1and 2 include the measured concentrations and imputed concentrations for the censored data, and the left panel

TABLE 2. Meta-Data from Studies and Derived BPA Concentrations in North American and European Surface Waters and Sediments

	North America		Europe	
	fresh	marine	fresh	marine
surface waters				
total number of weighted observations	1068	–	848	115
number of weighted observations < detection limit	852	–	415	58
percent observations < detection limit	80		49	50
number of imputed observations	1	–	10	0
median concentration (μg/L)	0.081	–	0.01	0.0016
95th percentile concentration (μg/L)	0.47	–	0.35	0.088
sediments				
total number of weighted observations	71	14	249	67
number of weighted observations < detection limit	32	11	75	44
percent observations < detection limit	45	79	30	66
number of imputed observations	35	–	65	0
median concentration (ng/g-dw)	0.6	3.5	16	8.5
90th percentile concentration (ng/g-dw)	3.4	ND[a]	162	207
95th percentile concentration (ng/g-dw)	ND	ND	256	566

[a] ND = Not determined.

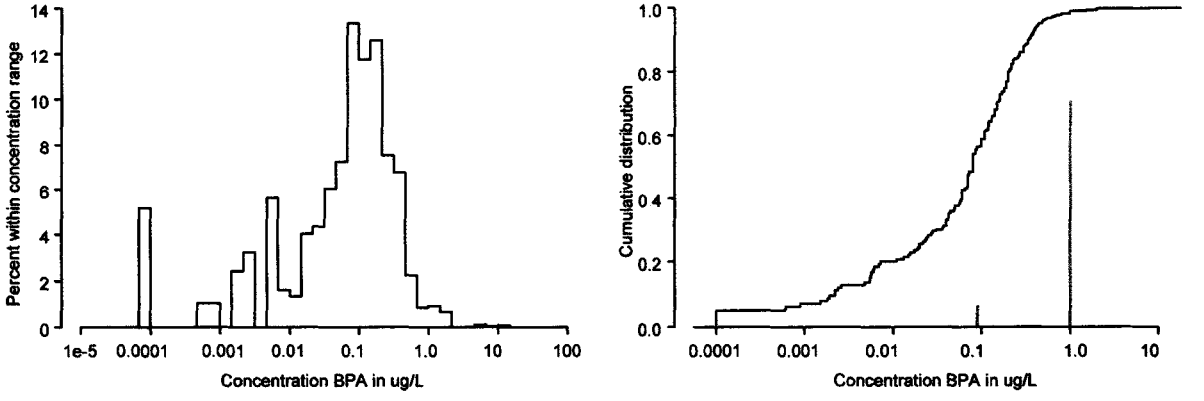

FIGURE 1. Concentration of BPA in surface water (fresh) in North America.

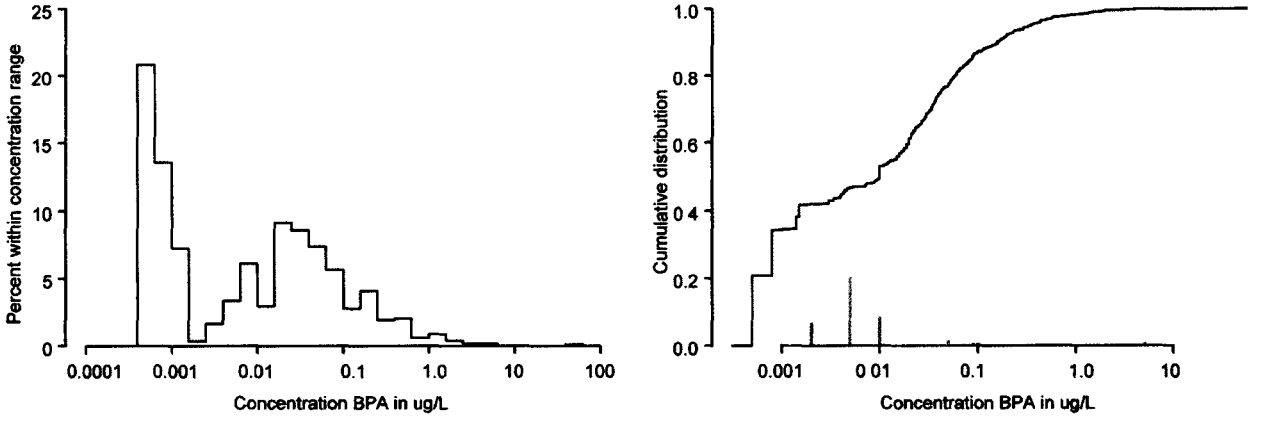

FIGURE 2. Concentration of BPA in surface water (fresh) in Europe.

presents the frequency histogram of the concentration. Each 10-fold increase in concentration is divided into size classes, where the number of classes was at most approximately the square root of the number of sampling locations. The right panels Figures 1 and 2 present the cumulative distribution. To illustrate the influence of variation in the detection limits and the fraction of measurements less than the detection limits, we have the right panels of Figures 1 and 2 showing, with vertical bars, the frequency of samples reported as less than the detection limit.

Concentration distributions for fresh and marine water are bimodal, i.e., there are a large number of samples with either a low measured concentration or a concentration less than a low detection limit. The distributions are positively skewed by a small number of measurements that reflect high concentrations. The highest concentration of BPA in North America (12 μg/L) was detected in a sample collected from the Santa Cruz River, AZ (*15*). Further communication with the author indicated that samples from this river were essentially 100% effluent from the local wastewater treatment plant (D.W. Kolpin, personal communication). BPA was also reported at 8 μg/L in the receiving waters from a manufacturing facility (U.S.A.), which was found to be composed of undiluted effluent (*16*). In Europe, BPA was reported at a

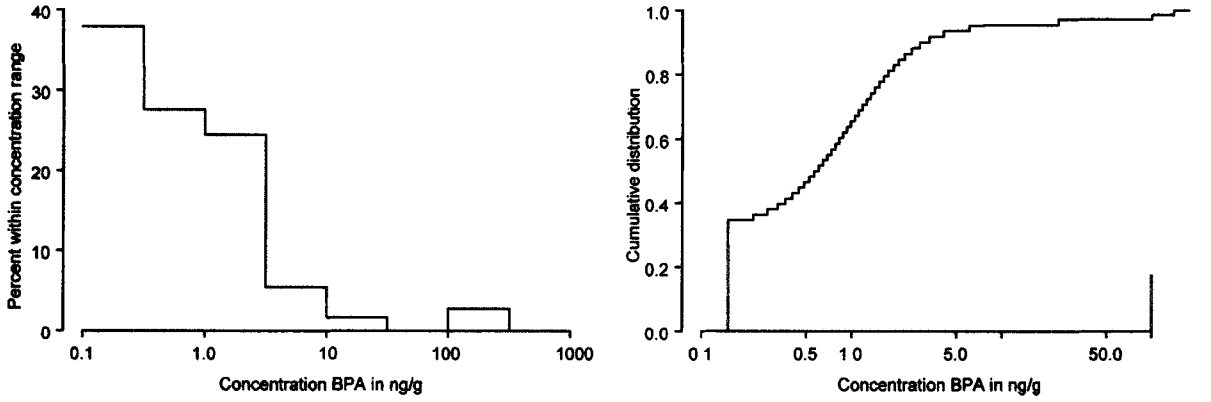

FIGURE 3. Concentration of BPA in freshwater sediment in North America.

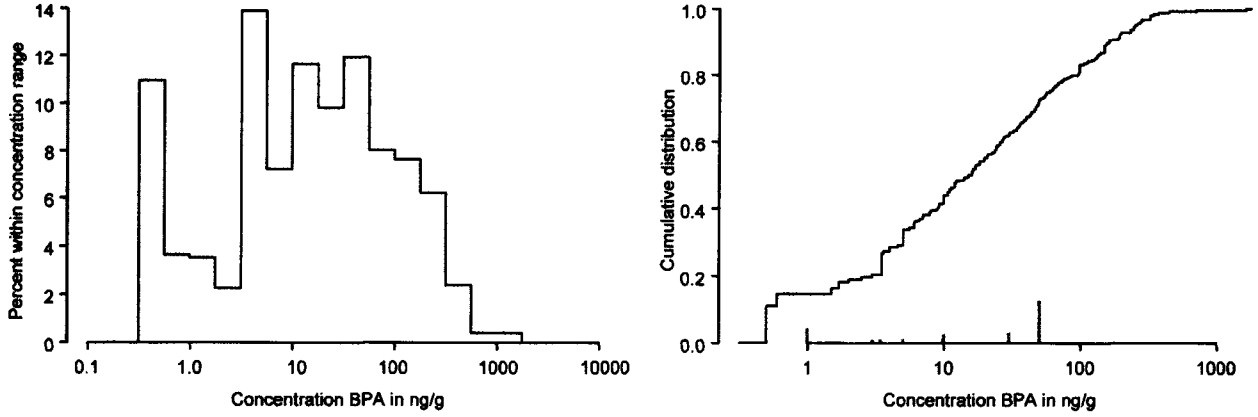

FIGURE 4. Concentration of BPA in freshwater sediment in Europe.

concentration of 43 μg/L in the Drammens River in Norway, which according to the authors is a site associated with industrial sources (*17*). Overall, the majority of BPA concentrations in fresh surface waters are low (<0.5 μg/L), with even lower concentrations in marine waters.

Sediment. In contrast to surface water, measurements of BPA concentrations in sediment are limited (Table 2). Three studies reporting measurements of BPA in freshwater sediment and one marine study were evaluated for North America, representing a combined total of 85 weighted observations. For Europe, a total of 17 studies reporting measurements of BPA in freshwater sediment (249 weighted observations) and 7 studies of marine sediment (67 weighted observations) were evaluated. The fraction of samples with concentrations less than the detection limit is smaller for freshwater sediments compared with those of marine sediments (Table 2, 45% versus 79% for North America and 30% versus 66% for Europe). Further, because of the lack of information on the individual data points, a larger percentage (approximately 25–50%) of the freshwater sediment samples were imputed for the calculations (35 for North America and 65 for Europe). The imputed observations fit a log-normal distribution and match the summary statistics available for that study. It was not necessary to estimate observations for the marine sediment calculations.

Table 2 summarizes the results of the statistical analysis of BPA in sediment. Concentration distributions for BPA in freshwater sediments in North America and Europe are shown in Figures 3 and 4 (additional distributions are presented in the Supporting Information). The median concentration in freshwater sediment in North America is less than that in Europe (0.6 versus 16 ng/g-dw). The 90th percentile concentration for North America of 3.4 ng/g-dw (the 95th percentile could not be calculated) is also less than the corresponding value of 162 ng/g-dw for Europe. The range of BPA concentrations for marine sediment in North America reported in the one available study was 1.5 to 5 ng/g-dw. For Europe, the 95th percentile values for fresh and marine sediments were 256 and 566 ng/g-dw, respectively. These results are in contrast to those for surface water, where the concentration of BPA was similar in North America and Europe. The differences likely reflect the limited availability of data for sediments and possibly the bias toward sampling sediments from locations that were suspected of contamination.

Discussion

This study was conducted to review the available monitoring data for North America and Europe and develop a statistical understanding of environmental exposures to BPA. The availability of measurements for BPA in surface water and sediment varies from one country to another. The purpose and scope of these studies also vary and include nationwide monitoring programs as well as those aimed at evaluating the influence of suspected contamination sources. In addition, among the national monitoring programs, the media evaluated are not always the same for one country compared to another. While these disparate studies make country-to-country comparisons impractical, the extent of data, particularly for surface water, provides a good indication of BPA concentrations on both continents.

The Kaplan–Meier method used in this evaluation was especially suited to the analysis of the results from multiple studies. The approach provided a means of resolving several key issues, including how to deal with the large amount of censored data and accommodating widely varying detection limits as well as providing a means to include studies where only summary statistics were available. Statistical analysis of the data suggested that median concentrations were better indicators of the central tendency of the data compared with

the means. Estimated mean concentrations were generally much larger than the medians due to the positively skewed distributions of the data that were affected by outliers in the data set. The 95th percentile concentrations were a better indicator of the upper limit concentrations than the maximum concentrations, which may be influenced by a single sample not representative of the majority of samples.

A large database was available for the evaluation of fresh surface waters for both continents. Given that data are available for 2682 total samples (1068 and 848 weighted observations for North America and Europe, respectively), we used a weight of evidence approach to develop an understanding of environmental exposures and their relationship to predicted no effect concentrations (PNEC) for aquatic organisms. Median BPA concentrations in freshwater in North America were higher than those for Europe (0.08 and 0.01 μg/L, respectively), although the 95th percentile concentrations were similar (0.47and 0.35 μg/L, respectively). Regional and local-scale surface water concentrations calculated for Europe using the EUSES model are reported in the European Union (EU) risk assessment report for BPA (*18*). The EUSES model relies on the assumption of dispersive emissions to the environment for regional-scale modeling and direct emission to surface water for local scenarios. Predicted freshwater and marine surface water concentrations for BPA are reported as ranging from 0.032 to 1.47 μg/L and 0.0027 to 0.13 μg/L, respectively. Overall, the predicted BPA concentrations for European surface waters are comparable to measured concentrations (*18*). Although not included in the present study, extensive monitoring data are also available for Japan (*19*). For surface water, a total of 3956 measurements have been obtained from 1120 sites from 752 rivers, lakes, and ponds. The concentration distribution of BPA in freshwater indicated that the BPA concentration at 30% of the surveyed sites was 0.005 μg/L or less, with the BPA concentration at 99% of the sites being 1 μg/L or less.

In contrast to the availability of monitoring data for surface waters, the data for sediments are more limited. Median and upper percentile BPA freshwater sediment concentrations were 0.6 and 3.4 ng/g-dw (90th) for North America versus 16 and 256 ng/g-dw (95th) for Europe, respectively. The median concentration of BPA in marine sediment in North America was 3.5 ng/g-dw (higher percentile values could not be calculated because of limited data). Median and upper 95th percentile concentrations of BPA in European marine sediments were 8.5 and 566 ng/g-dw, respectively. Predicted regional- and local-scale BPA concentrations for freshwater sediments reported in the EU risk assessment report ranged from 0.52 to 24 ng/g wet weight (1.35 to 62.4 ng/g-dw) (*18*), while predicted concentrations of BPA in marine sediments ranged from 0.034 to 2.1 ng/g wet weight (0.088 to 5.46 ng/g-dw). The difference between predicted and measured concentrations for Europe likely reflects the bias toward sampling sediments in areas susceptible to contamination.

For freshwater sediments, four countries in Europe have been studied more extensively than the rest. In Germany, 98 samples were collected from 1996 to 2000; many of these samples were taken as part of the Arge-Elbe program (*20–22*). The 95th percentile concentrations ranged from 13 to 311 ng/g-dw. In the United Kingdom, 65 samples were collected between 1999 and 2003 and include those from a nationwide sediment monitoring program (*23*). The frequency of detection ranged from 4% to 53%, and the maximum concentration reported was 56.8 ng/g-dw. Sweden has also reported results of a nationwide sediment monitoring program (*24*). Of the 35 samples collected from 2003 to 2004, BPA was detected in only 9% of the samples; the 95th percentile concentration was 83 ng/g-dw. The levels of BPA in Norwegian sediments have been reported in a number of studies. Fjeld et al. (*25, 26*) described BPA concentrations in heavily industrialized regions or contaminated sites. Elsewhere, 95th percentile concentrations of BPA were about 50 ng/g-dw. The findings for the nonindustrialized areas are in good agreement with a recently published Norwegian report that appeared too late to be included in the statistical analysis (*27*). In the recent study, BPA concentrations in sediment samples collected from 26 stations were typically between approximately 3 and 15 ng/g-dw. Two elevated concentrations of 93 and 106 ng/g-dw were associated with surface runoff near urban areas.

The highest concentrations of BPA in European sediments were typically found in areas that were suspected of being impacted because of their proximity to industrial activity. For example, maximum BPA sediment concentrations in the Elbe River ranged from 322 to 1630 ng/g-dw (*20, 22, 28*). According to the authors, many of the regions of the Elbe River that were sampled are highly industrialized and receive discharges from numerous municipal and industrial treatment plants. In Norway, Fjeld et al. (*25*) reported sediment concentrations in samples taken from industrially contaminated sites (371.16 ng/g-dw) and from the Drammens River (industrialized area, 279.8 ng/g-dw).

Because the details were not available, the results of a United States nationwide soil and sediment reconnaissance (*29*) were not included in the present analysis. According to the authors, analytical methods were developed for 61 compounds and applied to a set of 103 environmental soil, sediment, and suspended sediment samples collected from urban sites throughout the United States. BPA was detected above the detection limit (32 ng/g-dw) in only 20 of the 103 samples. The 50th and 90th percentiles for the measured concentrations were approximately 100 and 400 ng/g-dw, respectively.

To assess the potential ecological significance, we compared exposure concentrations for North America and Europe with available regulatory criteria. For example, the European Chemicals Bureau (*18*) has recently completed a comprehensive risk assessment for BPA and has defined predicted no effect concentrations (PNEC) for fresh and marine waters and sediment. The PNEC for freshwater and marine waters are 1.5 and 0.15 μg/L, respectively. Comparison of the measured concentrations summarized in Table 2 indicates that all values are below the PNEC values, suggesting a low potential for adverse effects on aquatic ecosystems. Similar conclusions were reached in a comprehensive risk assessment conducted in Japan (*19*). In contrast, on the basis of a less comprehensive screening assessment, which included studies considered not relevant elsewhere (*18*), Environment Canada has recently concluded concentrations entering the Canadian environment may pose a risk to aquatic organisms and suggested further monitoring is appropriate (*30*).

The European risk assessment (*18*) also calculated PNEC for freshwater and marine sediments of 63 and 6.3 ng/g-dw, respectively. All predicted concentrations in the European risk assessment (*18*) were below the PNEC. However, comparison of the measured concentrations summarized in Table 2 indicates that while 90th percentile concentrations in North American sediment are below the PNEC, the 95th percentile concentrations in European fresh and marine sediments exceed the PNEC. These results further reflect the emphasis that was made in the selection of sites in sampling and analysis programs in Europe toward highly urbanized and industrialized locations that receive municipal and industrial waste treatment discharges.

The approaches outlined in this study provide a statistical basis for understanding environmental exposures. As additional monitoring data become available, they can be compared to the database developed in this study. Median and upper 90th or 95th percentile concentrations measured in North American fresh and marine waters and sediments were all below the PNEC developed for the European risk

assessment. Measured and predicted concentrations of BPA in European surface waters and predicted concentrations in European sediments were all below their respective PNEC. Measured concentrations of BPA in European sediments that exceed the PNEC were generally associated with wastewater effluent dominated streams and heavily urbanized or industrialized locations.

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Supporting Information Available

Tables providing details for all studies used in this analysis and additional concentration distributions. This material is available free of charge via the Internet at http://pubs.acs.org.

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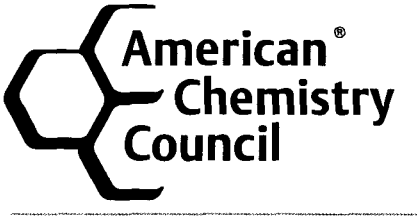

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